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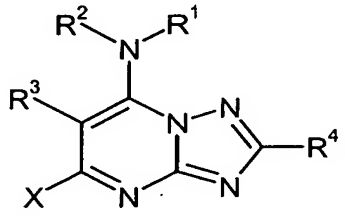
Triazolopyrimidines

The invention relates to triazolopyrimidines, to a process for their preparation and to their use for controlling unwanted microorganisms.

It is already known that certain triazolopyrimidines have fungicidal properties (cf. EP 0 550 113-A, WO 94-20 501, EP 0 613 900-A, US 5 612 345-A, EP 0 834 513-A, WO 98-46 607 and WO 98-46 608). The activity of these compounds is good; however, at low application rates it is sometimes unsatisfactory.

However, since the ecological and economical demands made on modern fungicides are increasing constantly, for example with respect to activity spectrum, toxicity, selectivity, application rate, formation of residues and favorable manufacture, and there can furthermore be problems, for example, with resistance, there is a constant need to develop novel fungicides which, at least in some areas, have advantages over those of the prior art.

This invention now provides novel triazolopyrimidines of the formula



(I)

in which

R^1 represents H, R^2, optionally substituted alkyl, optionally substituted alkenyl, optionally substituted alkynyl, optionally substituted cyloalkyl or represents optionally substituted heterocyclyl;

R^2 represents an organic radical which contains 3 to 13 carbon atoms and one or more silicon atoms and, if appropriate, 1 to 3 identical or different heteroatoms from the group consisting of oxygen, nitrogen and sulfur, and which is unsubstituted or substituted by 1 to 4 identical or different halogens; or

R^1 and R^2 together with the nitrogen atom to which they are attached represent an optionally substituted heterocyclic ring which contains one or more silicon atoms and/or is substituted by one or more radicals R^2;

R^3 represents optionally substituted aryl, optionally substituted heterocyclyl, optionally substituted alkyl, optionally substituted alkenyl, optionally substituted alkynyl, optionally

substituted cycloalkyl, optionally substituted aralkyl, or optionally substituted amino group, optionally substituted (C_1-C_8)-alkoxy, optionally substituted (C_1-C_8)-alkylthio, optionally substituted (C_6-C_{10})-aryloxy, optionally substituted (C_6-C_{10})-arylthio, optionally substituted heterocyclyloxy, optionally substituted heterocyclyloxy, optionally substituted C_6-C_{10})-aryl-(C_1-C_4)-alkoxy, optionally substituted (C_6-C_{10})-aryl-(C_1-C_4)-alkylthio, optionally substituted heterocyclyl-(C_1-C_4)-alkoxy, or optionally substituted heterocyclyl-(C_1-C_4)-alkylthio;

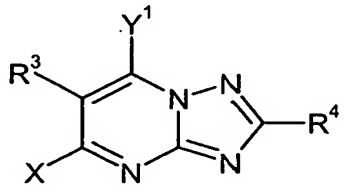
R^4 represents H, halogen, optionally halogen-substituted alkyl or optionally halogen-substituted cycloalkyl and

X represents halogen, cyano, optionally substituted alkyl, optionally substituted alkoxy, optionally substituted alkythio, optionally substituted alkylsulfinyl, optionally substituted alkylsulfonyl or optionally substituted phenyl,

and their salts.

Furthermore, it has been found that triazolopyrimidines of the formula (I) can be prepared by

(a) reacting halotriazolopyrimidines of the formula



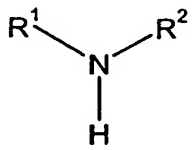
(II)

in which

R^3, R^4 and X are as defined above and

Y^1 represents halogen,

with amines of the formula



(III)

in which

R^1 and R^2 are as defined above,

if appropriate in the presence of a diluent, if appropriate in the presence of an acid acceptor and if appropriate in the presence of a catalyst.

Finally, it has been found that the triazolopyrimidines of the formula (I) are highly suitable for controlling unwanted microorganisms. Especially, they have strong fungicidal activity and can be used both in crop protection and in the protection of materials.

The compounds according to the invention of formula (I) can, if appropriate, be present as mixtures of different possible isomeric forms, in particular stereoisomers, such as E and Z, threo and erythro, and also optical isomers, such as R and S isomers or atrope isomers, and, if appropriate, also of tautomers.

The formula (I) provides a general definition of the triazolopyrimidines according to the invention.

Preference is given to compounds of the formula (I) in which

a^1) R^3 represents optionally substituted aryl, or

a^2) R^3 represents optionally substituted heterocyclyl, or

a^3) R^3 represents optionally substituted alkyl, or

a^4) R^3 represents optionally substituted alkenyl, or

a^5) R^3 represents optionally substituted alkynyl, or

a^6) R^3 represents optionally substituted cycloalkyl, or

a^7) R^3 represents optionally substituted aralkyl, or

a^8) R^3 represents an optionally substituted amino group.

Preference is likewise given to compounds of the formula (I) which R^3 has one of the meanings below:

b^1: $a^1, a^2, a^3, a^4, a^5, a^6, a^7,$

b^2: $a^1, a^2, a^3, a^4, a^5, a^6, a^8,$

b^3: $a^1, a^2, a^3, a^4, a^5, a^7, a^8,$

b^4: $a^1, a^2, a^3, a^4, a^6, a^7, a^8,$

b^5: a^1, a^2, a^3, a^5, a^6, a^7, a^8,

b^6: a^1, a^2, a^4, a^5, a^6, a^7, a^8,

b^7: a^1, a^3, a^4, a^5, a^6, a^7, a^8,

b^8: a^2, a^3, a^4, a^5, a^6, a^7, a^8.

Preference is furthermore given to those compounds of the formula (I) in which one or more symbols have one of the preferred meanings given below, i.e. in which

R^1 represents H, or

R^1 represents a radical R^2, or

R^1 represents alkyl having 1 to 6 carbon atoms which may be mono- to pentasubstituted by identical or different substituents from the group consisting of halogen, cyano, hydroxy, alkoxy having 1 to 4 carbon atoms and cycloalkyl having 3 to 8 carbon atoms, or

R^1 represents alkenyl having 2 to 6 carbon atoms which may be mono- to trisubstituted by identical or different substituents from the group consisting of halogen, cyano, hydroxy, alkoxy having 1 to 4 carbon atoms and cycloalkyl having 3 to 8 carbon atoms, or

R^1 represents alkynyl having 3 to 6 carbon atoms which may be mono- to trisubstituted by identical or different substituents from the group consisting of halogen, cyano, alkoxy having 1 to 4 carbon atoms and cycloalkyl having 3 to 8 carbon atoms, or

R^1 represents cycloalkyl having 3 to 8 carbon atoms which may be mono- to trisubstituted by identical or different substituents from the group consisting of halogen and alkyl having 1 to 4 carbon atoms, or

R^1 represents saturated or unsaturated heterocyclyl having 3 to 8 ring members and 1 to 3 heteroatoms, such as nitrogen, oxygen and/or sulfur, where the heterocyclyl may be mono- or disubstituted by halogen, alkyl having 1 to 4 carbon atoms, cyano and/or cycloalkyl having 3 to 8 carbon atoms,

R^2 represents an aliphatic saturated or unsaturated group having 1 to 13 carbon atoms and one or more silicon atoms which optionally contains 1 to 3 identical or different heteroatoms from the group consisting of oxygen, sulfur and nitrogen and which is unsubstituted or substituted by 1 to 4 identical or different halogen atoms, or

R^1 and R^2 together with the nitrogen atom to which they are attached represent a saturated or unsaturated heterocyclic ring having 3 to 8 ring members which contains one or more silicon atoms and/or is substituted by one or more radicals R^2, where the heterocycle may contain a further nitrogen, oxygen or sulfur atom as ring member and where the heterocycle may furthermore be substituted up to three times by fluorine, chlorine, bromine, alkyl having 1 to 4 carbon atoms and/or haloalkyl having 1 to 4 carbon atoms and 1 to 9 fluorine and/or chlorine atoms,

R^3 represents C_1-C_{10}-alkyl, C_2-C_{10}-alkenyl, C_2-C_{10}-alkynyl, C_3-C_8-cycloalkyl, phenyl-C_1-C_{10}-alkyl where R^3 is unsubstituted or partly or fully halogenated and/or optionally carries one to three radicals from the group R^X, or C_1-C_{10}-halogenalkyl which optionally carries one to three radicals from the group R^X, and R^X represents cyano, nitro, hydroxy, C_1-C_6-alkyl, C_1-C_6-haloalkyl, C_3-C_6-cycloalkyl, C_1-C_6-alkoxy, C_1-C_6-haloalkoxy, C_1-C_6-alkylthio, C_1-C_6-halogenalkylthio, C_1-C_6-alkylsulfinyl, C_1-C_6-halogenalkyl-sulfinyl, C_1-C_6-alkylsulfonyl, C_1-C_6-halogenalkylsulfonyl, C_1-C_6-alkylamino, di-C_1-C_6-alkylamino, C_2-C_6-alkenyl, C_2-C_6-alkenyloxy, C_2-C_6-alkynyl, C_3-C_6-alkynyloxy and optionally halogenated oxy-C_1-C_4-alkyl-C_1-C_4-alkeneoxy, oxy-C_1-C_4-alkenyl-C_1-C_4-alkoxy, oxy-C_1-C_4-alkyl-C_1-C_4-alkyloxy,

R^3 represents phenyl which may be mono- to tetrasubstituted by identical or different substituents from the group consisting of

halogen, cyano, nitro, amino, hydroxy, formyl, carboxy, carbamoyl, thiocarbamoyl;

in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulfinyl or alkylsulfonyl having in each case 1 to 6 carbon atoms;

in each case straight-chain or branched alkenyl or alkenyloxy having in each case 2 to 6 carbon atoms;

in each case straight-chain or branched haloalkyl, haloalkoxy, haloalkylthio, haloalkylsulfinyl or haloalkylsulfonyl having in each case 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms;

in each case straight-chain or branched haloalkenyl or haloalkenyloxy having in each case 2 to 6 carbon atoms and 1 to 11 identical or different halogen atoms;

in each case straight-chain or branched alkylamino, dialkylamino, alkylcarbonyl, alkyl-carbonyloxy, alkoxycarbonyl, alkylsulfonyloxy, hydroximinoalkyl or alkoximinoalkyl having in each case 1 to 6 carbon atoms in the individual alkyl moieties;

cycloalkyl having 3 to 8 carbon atoms;

2,3-attached 1,3-propanediyl, 1,4-butanediyl, methylenedioxy (-O-CH_2-O-) or 1,2-ethylenedioxy (-O-CH_2-CH_2-O-), where these radicals may be mono- or polysubstituted by identical or different substituents from the group consisting of halogen, alkyl having 1 to 4 carbon atoms and haloalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms;

or

R^3 represents saturated or unsaturated heterocyclyl having 3 to 8 ring members and 1 to 3 heteroatoms from the group consisting of nitrogen, oxygen and sulfur, where the heterocyclyl may be mono- or disubstituted by halogen, alkyl having 1 to 4 carbon atoms, alkoxy having 1 to 4 carbon atoms, alkylthio having 1 to 4 carbon atoms, haloalkoxy having 1 to 4 carbon atoms, haloalkylthio having 1 to 4 carbon atoms, cyano, nitro and/or cycloalkyl having 3 to 6 carbon atoms;

or

R^3 represents C_1-C_8-alkylamino, C_2-C_8-alkenylamino, C_2-C_8-alkynylamino, di-C_1-C_8-alkylamino, di-C_2-C_8-alkenylamino, di-C_2-C_8-alkynylamino, C_2-C_8-alkenyl-(C_2-C_8)-alkynylamino, C_2-C_6-alkynyl-(C_1-C_8)-alkylamino, C_2-C_8-alkenyl-(C_1-C_8)-alkylamino, C_6-C_{10}-arylamino, C_6-C_{10}-aryl-(C_1-C_8)-alkylamino, C_6-C_{10}-aryl-(C_1-C_4)-alkyl-(C_1-C_8)-alkylamino, heterocyclyl-(C_1-C_8)-alkylamino or heterocyclyl-(C_1-C_4)-alkyl-(C_1-C_8)-alkylamino;

R^4 represents H, halogen, (C_1-C_4)-alkyl which is unsubstituted or substituted by one or more halogen atoms, cyclopropyl which is unsubstituted or substituted by one or more halogen atoms, and

X represents fluorine, chlorine, bromine, CN, (C_1-C_4)-alkyl which is unsubstituted or substituted by one or more fluorine or chlorine atoms, (C_1-C_4)-alkoxy which is unsubstituted or substituted by one or more fluorine or chlorine atoms or (C_1-C_4)-alkylthio which is unsubstituted or substituted by one or more fluorine or chlorine atoms.

Particular preference is given to those triazolopyrimidines of the formula (I) in which one or more symbols have one of the particularly preferred meanings listed below, i.e. in which

R^1 represents hydrogen, methyl or ethyl, or

R^2 represents a group of the formula Y^2-$Si(O_mCH_3)(O_nCH_3)(O_pY^3)$,

where m, n and p independently of one another represent 0 or 1;

Y^2 represents a bond or alkanediyl, alkenediyl or alkynediyl, each of which is straight-chain or branched, has 1 to 6 or 2 to 6 carbon atoms, is optionally interrupted by one or two nonadjacent oxygen atoms and is unsubstituted or substituted by one to three identical or different halogen atoms;

Y^3 represents straight-chain or branched alkyl or alkenyl having 1 to 5 or 2 to 5 carbon atoms, optionally interrupted by an oxygen-nitrogen or sulfur atom and unsubstituted or substituted by 1 to 3 identical or different halogen atoms;

R^3 represents $(C_1$-$C_8)$-alkyl, $(C_1$-$C_8)$-cycloalkyl or benzyl or

R^3 represents phenyl which may be mono- to trisubstituted by identical or different substituents from the group consisting of

fluorine, chlorine, bromine, cyano, nitro, formyl, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, allyl, propargyl, methoxy, ethoxy, n- or i-propoxy, methylthio, ethylthio, n- or i-propylthio, methylsulfinyl, ethylsulfinyl, methylsulfonyl, ethylsulfonyl, allyloxy, propargyloxy, trifluoromethyl, trifluoroethyl, difluoromethoxy, trifluoromethoxy, difluorochloromethoxy, trifluoroethoxy, difluoromethylthio, difluorochloromethylthio, trifluoromethylthio, trifluoromethylsulfinyl, trifluoromethylsulfonyl, trichloroethynyloxy, trifluoroethynyloxy, chloroallyloxy, iodopropargyloxy, methylamino, ethylamino, n- or i-propylamino, dimethylamino, diethylamino, acetyl, propionyl, acetyloxy, methoxycarbonyl, ethoxycarbonyl, hydroximinomethyl, hydroximinoethyl, methoximinomethyl, ethoximinomethyl, methoximinoethyl, ethoximinoethyl, cyclopropyl, cyclobutyl, cyclopentyl or cyclohexyl,

2,3-attached 1,3-propanediyl, 1,4-butanediyl, methylenedioxy (-O-CH_2-O-) or 1,2-ethylenedioxy (-O-CH_2-CH_2-O-), where these radicals may be mono- or polysubstituted by identical or different substituents from the group consisting of fluorine, chlorine, methyl, ethyl, n-propyl, i-propyl and trifluoromethyl.

R^3 represents pyridyl which is attached in the 2- or 4-position and may be mono- to tetrasubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, methoxy, methylthio, hydroximinomethyl, hydroximinoethyl, methoximinomethyl, methoximinoethyl and trifluoromethyl, or

R^3 represents pyrimidyl which is attached in the 2- or 4-position and may be mono- to trisubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, methoxy, methylthio, hydroximinomethyl, hydroximinoethyl, methoximinomethyl, methoximinoethyl and trifluoromethyl, or

R^3 represents thienyl which is attached in the 2- or 3-position and may be mono- to trisubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, methoxy, methylthio, hydroximinomethyl, hydroximinoethyl, methoximinomethyl, methoximinoethyl and trifluoromethyl, or

R^3 represents C_1-C_8-alkylamino or di-C_1-C_8-alkylamino, or

R^3 represents thiazolyl which is attached in the 2-, 4- or 5-position and may be mono- to trisubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, methoxy, methylthio, hydroximinomethyl, hydroximinoethyl, methoximinomethyl, methoximinoethyl and trifluoromethyl, or

R^3 represents N-piperidinyl, N-tetrazolyl, N-pyrazolyl, N-imidazolyl, N-1,2,4-triazolyl, N-pyrrolyl, or N-morpholinyl, each of which is unsubstituted or mono- or - if possible – polysubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, methoxy, methylthio, hydroximinomethyl, hydroximinoethyl, methoximinomethyl, methoximinoethyl and trifluoromethyl,

R^4 represents H, Cl, F, CH_3, $-CH(CH_3)_2$ or cyclopropyl; and

X represents F, Cl, CN, (C_1-C_4)-alkyl which is unsubstituted or substituted by one or more fluorine or chlorine atoms, OCH_3 or SCH_3.

Very particular preference is given to compounds of the formula (I) in which one or more of the symbols have one of the very particularly preferred meanings listed below, in which

R^1 represents H;

R^2 represents $SiMe_3$, $SiMe_2Et$, $SiMe_2CHMe_2$, $SiMe_2CH_2CHMe_2$, $SiMe_2CH_2CMe_3$, $SiMe_2OCHMe_2$, $SiMe_2OCH_2CHMe_2$, CH_2SiMe_3, CH_2SiMe_2Et, $CH_2SiMe_2CHMe_2$, $CH_2SiMe_2CH_2CHMe$, CH_2SiMe_2OMe, $CH_2SiMe_2OCHMe_2$, $CH_2SiMe_2OCH_2CHMe_2$, $CHMeSiMe_3$, $CHMeSiMe_2OMe$, $(CH_2)_2SiMe_3$, $(CH_2)_2SiMe_2Et$, $(CH_2)_2SiMe_2CHMe_2$, $(CH_2)_2SiMe_2CMe_3$, $(CH_2)_2SiMe_2CH_2CHMe_2$, $(CH_2)_2SiMe_2CH_2CH_2Me$, $(CH_2)_2SiMe_2CH_2CMe_3$, $(CH_2)_2SiMe_2OCHMe_2$, $(CH_2)_2SiMe_2OCH_2CHMe_2$, $CHMeCH_2SiMe_3$, $CHMeCH_2SiMe_2Et$, $CHMeCH_2SiMe_2CH_2CH_2Me$,

$CHMeCH_2SiMe_2CHMe_2$, $CHMeCH_2SiMe_2CMe_3$, $CHMeCH_2SiMe_2CH_2CHMe_2$, $CFMeCH_2SiMe_3$, $CHMeCH_2CH_2SiMe_2OMe$, $CHMeCH_2SiMe_2OCHMe_2$, $CHMeCH_2SiMe_2OCH_2CHMe_2$, $CH_2CHMeSiMe_3$, $CH_2CHMeSiMe_2Et$, $CH_2CHMeSiMe_2CHMe_2$, $CHMeCHMeSiMe_3$, $CMe_2CH_2SiMe_3$, $(CH_2)_3SiMe_3$, $(CH_2)_3SiMe_2Et$, $(CH_2)_3SiMe_2CHMe_2$, $(CH_2)_3SiMe_2CH_2CHMe_2$, $(CH_2)_3SiMe_2OMe$, $(CH_2)_3SiMe_2OCHMe_2$, $(CH_2)_3SiMe_2OCH_2CHMe_2$, $CHMeCH_2CH_2SiMe_3$, $CHMeCH_2CH_2SiMe_2Et$, $CHMeCH_2CH_2SiMe_2CHMe_2$, $CHMeCH_2CH_2CH_2SiMe_2OMe$, $CHMeCH_2CH_2SiMe_2OCHMe_2$, $CMe{=}CHSiMe_3$, $CH_2CH_2SiMe_2OMe$, $-C{\equiv}C-SiMe_3$, $-CH_2-C{\equiv}C-SiMe_3$ or $-CHMe-C{\equiv}C-SiMe_3$;

R^3 represents $(C_1\text{-}C_6)$-alkyl, $(C_3\text{-}_6)$-alkenyl, $(C_3\text{-}C_6)$-alkynyl, $(C_3\text{-}C_8)$-cycloalkyl, where R^3 is unsubstituted or substituted by one or more fluorine or chlorine atoms,

or

R^3 represents 2,4- or 2,6-disubstituted phenyl, or represents 2-substituted phenyl or represents 2,4,6-trisubstituted phenyl,

R^3 represents pyridyl which is attached in the 2- or 4-position and may be mono- to tetrasubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine, cyano, methyl, ethyl, methoxy, methylthio, hydroximinomethyl, hydroximinoethyl, methoximinomethyl, methoximinoethyl and trifluoromethyl, or

R^3 represents pyrimidyl which is attached in the 4-position and may be mono- to trisubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine, cyano, methyl, ethyl, methoxy, methylthio, hydroximinomethyl, hydroximinoethyl, methoximinomethyl, methoximinoethyl and trifluoromethyl;

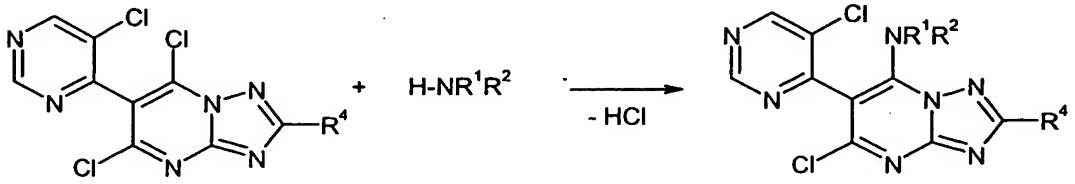
R^4 represents H, $-CH_3$, $-CH(CH_3)_2$, Cl or cyclopropyl,

and

X represents fluorine, chlorine, CN, $(C_1\text{-}C_3)$-alkyl, in particular CH_3 or $(C_1\text{-}C_3)$-haloalkyl, in particular CF_3, OCH_3, or SCH_3.

The radical definitions mentioned above can be combined with one another as desired. Moreover, individual definitions may not apply.

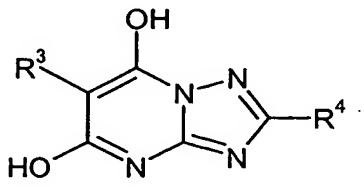
Using, for example, 5,7-dichloro-6-(5-chloropyrimidin-4-yl)-[1,2,4]triazolo[1,5-a]pyrimidine as starting material, the course of the process (a) according to the invention can be illustrated by the formula scheme below.



The formula (II) provides a general definition of the dihalotriazolopyrimidines required as starting materials for carrying out the process (a) according to the invention. In this formula (II), R^3 and X preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred for these radicals. Y^1 preferably represents fluorine, chlorine or bromine, particularly preferably fluorine or chlorine.

Dihalotriazolopyrimidines can be prepared by reacting, for example,

(b) dihydroxytriazolopyrimidines of the formula



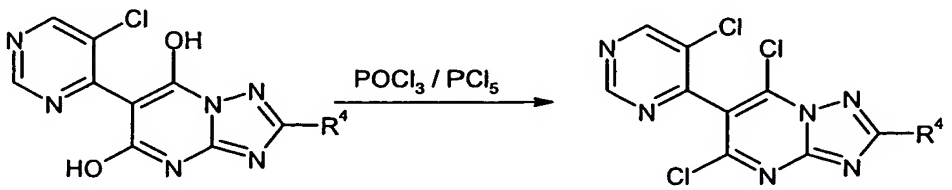
(IV)

in which

R^3 is as defined above,

with halogenating agents, if appropriate in the presence of a diluent.

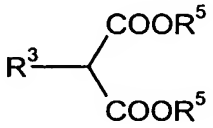
Using, for example, 6-(5-chloropyrimidin-4-yl)-[1,2,4]triazolo[1,5-a]-pyrimidine-5,7-diol as starting material and phosphorus oxychloride in a mixture with phosphorus pentachloride as halogenating agent, the course of the process (b) according to the invention can be illustrated by the formula scheme below.



The formula (IV) provides a general definition of the dihydroxytriazolopyrimidines required as starting materials for carrying out the process (b). In this formula, R^3 preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred for this radical.

Dihydroxytriazolopyrimidines of the formula (IV) can be prepared by reacting, for example,

(c) heteroarylmalonic esters of the formula



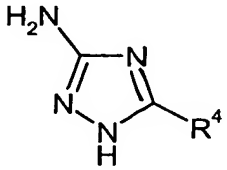
(V)

in which

R^3 is as defined above and

R^5 represents alkyl having 1 to 4 carbon atoms,

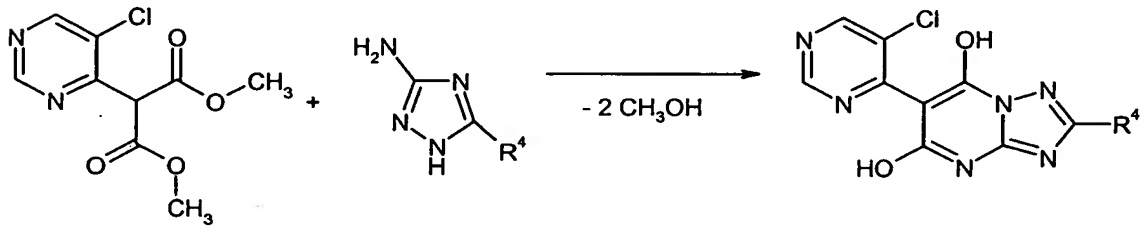
with an aminotriazole of the formula



(VI)

if appropriate in the presence of a diluent and if appropriate in the presence of an acid binder.

Using, for example, dimethyl 2-(5-chloropyrimidin-4-yl)malonate and a 3-aminotriazole as starting materials, the course of the process (c) according to the invention can be illustrated by the formula scheme below.

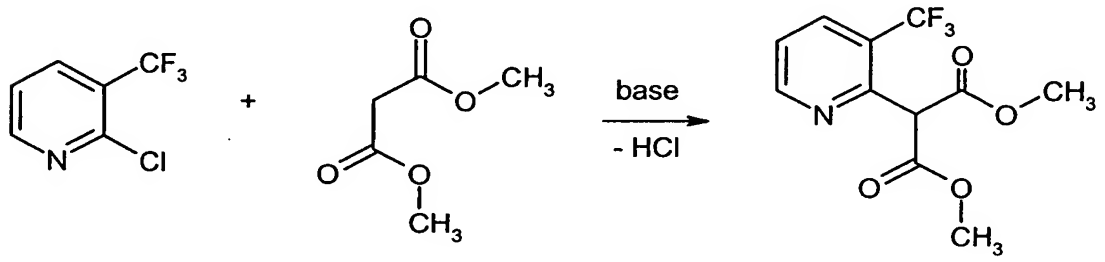


The formula (V) provides a general definition of the heteroarylmalonic esters required as starting materials for carrying out the process (c) according to the invention. In this formula, R^3 preferably has those meanings which have already been mentioned in connection with the description of the

compounds of the formula (I) according to the invention as being preferred for this radical. R^5 represents methyl or ethyl.

The heteroarylmalonic esters of the formula (V) are known (cf. DE 38 20 538-A, WO 01-11 965 and DE-A 103 25 133).

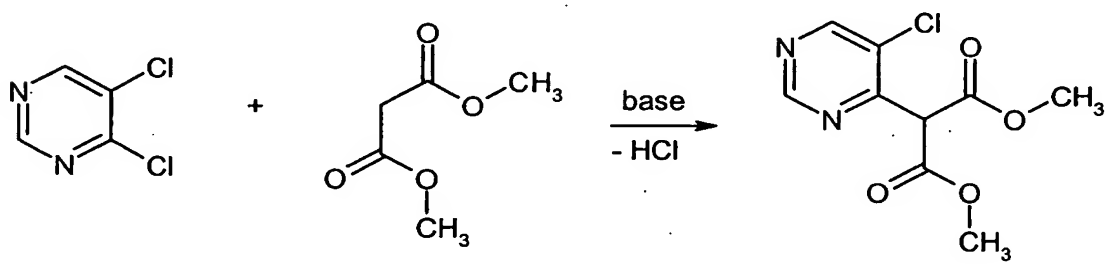
Using, for example, 2-chloro-3-trifluoromethylpyridine and dimethyl malonate as starting materials, the course of the process (d) according to the invention can be illustrated by the formula scheme below.



The halopyridines required as starting materials for carrying out the process (d) according to the invention are known chemicals for synthesis.

The malonic esters furthermore required as starting materials for carrying out the process (d) according to the invention are likewise known chemicals for synthesis.

Using, for example, 4,5-dichloropyrimidine and dimethyl malonate as starting materials, the course of the process (e) according to the invention can be illustrated by the formula scheme below.



The halopyrimidines required as starting materials for carrying out the process (e) according to the invention are known and can be prepared with known methods (cf. J. Chem. Soc. 1955, 3478, 3481).

The aminotriazoles of the formula (VI) furthermore required as starting material for carrying out the process (c) according to the invention are commercial chemicals.

Suitable halogenating agents for carrying out the process (b) are all components customary for replacing hydroxyl groups by halogen. Preference is given to using phosphorus trichloride, phosphorus tribromide, phosphorus pentachloride, phosphorus oxychloride, thionyl chloride, thionyl bromide, phosgene, diphosgene, triphosgene or mixtures thereof, in particular a mixture of phosphorus oxychloride, phosphorus trichloride and chlorine (see, for example, EP-A 1 077 210). The corresponding fluorine compounds of the formula (II) can be prepared from the chlorine or bromine compounds by reaction with potassium fluoride.

The halogenating agents mentioned are known.

The formula (III) provides a general definition of the amines required as starting materials for carrying out the process (a) according to the invention. In this formula, R^1 and R^2 preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred for R^1 and R^2.

The synthesis of compounds of the formula (II) in which R^3 represents an alkyl, alkenyl, alkynyl or cycloalkyl group is described in WO-A 03/009687.

The synthesis of compounds of the formula (II) in which R^3 carries an amino, alcohol or thio function is described in WO-A 03/039 259.

The amines of the formula (III) are known. Some of them are commercially available, or they can be prepared by known methods familiar to the person skilled in the art.

Thus, silylated amines of the formula (IIIa)

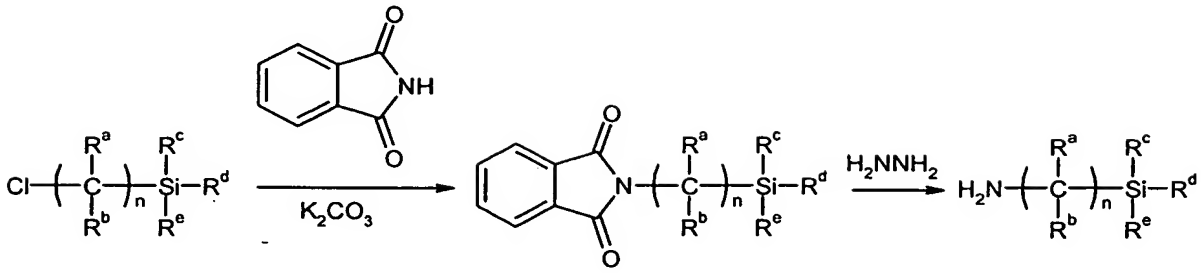
$$H_2N\text{-}(CR^aR^b)_n\text{-}SiR^cR^dR^e$$

in which

n is a natural number from 0 to 10 and

R^a, R^b, R^c, R^d are identical or different and H, CH_3 or C_2H_5 (the total number of carbon atoms in R^{a-d} being ≤ 12),

are generally obtainable, for example by reacting phthalimide in the presence of a base, such as K_2CO_3, with a haloalkylsilane and cleaving the resulting N-substituted phthalimide with hydrazine:



Such syntheses are described, for example, in J. Am. Chem. Soc. 1951, 73, 5130 or J. Organomet. Chem. 1978, 174, C18.

Haloalkylsilanes are commercially available or can be prepared by known methods familiar to the person skilled in the art (see, for example, Houben-Weyl, volume 13/5, p. 65 ff. or Science of Synthesis, vol. 4, p. 247 ff.).

Suitable diluents for carrying out the process (a) according to the invention are all customary inert organic solvents. Preference is given to using halogenated hydrocarbons, such as, for example, chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride, dichloroethane or trichloroethane; ethers, such as diethyl ether, diisopropyl ether, methyl t-butyl ether, methyl t-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole; nitriles, such as acetonitrile, propionitrile, n- or i-butyronitrile or benzonitrile; amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide; esters, such as methyl acetate or ethyl acetate; sulfoxides, such as dimethyl sulfoxide; sulfones, such as sulfolane.

Suitable acid acceptors for carrying out the process (a) according to the invention are all inorganic or organic bases customary for such reactions. Preference is given to using alkaline earth metal or alkali metal hydrides, hydroxides, amides, alkoxides, acetates, carbonates or bicarbonates, such as, for example, sodium hydride, sodium amide, lithium diisopropylamide, sodium methoxide, sodium ethoxide, potassium tert-butoxide, sodium hydroxide, potassium hydroxide, sodium acetate, potassium acetate, calcium acetate, sodium carbonate, potassium carbonate, potassium bicarbonate and sodium bicarbonate, and furthermore ammonium compounds such as ammonium hydroxide, ammonium acetate and ammonium carbonate, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, N,N-dimethylbenzylamine, pyridine, N-methylpiperidine, N-methylmorpholine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

Suitable catalysts for carrying out the process (a) according to the invention are all reaction promoters customary for such reactions. Preference is given to using fluorides, such as sodium fluoride, potassium fluoride or ammonium fluoride.

When carrying out the process (a) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the process is carried out at temperatures between 0°C and 150°C, preferably at temperatures between 0°C and 80°C.

When carrying out the process (a) according to the invention, in general from 0.5 to 10 mol, preferably from 0.8 to 2 mol, of amine of the formula (III) are employed per mole of dihalotriazolopyrimidine of the formula (II). Work-up is carried out by customary methods.

Suitable diluents for carrying out the process (b) according to the invention are all solvents customary for such halogenations. Preference is given to using halogenated aliphatic or aromatic hydrocarbons, such as chlorobenzene. However, it is also possible for the halogenating agent itself, for example phosphorus oxychloride, or a mixture of halogenating agents to act as diluent.

When carrying out the process (b), the temperatures can be also be varied within a relatively wide range. In general, the process is carried out at temperatures between 0°C and 150°C, preferably between 10°C and 120°C.

When carrying out the process (b), dihydroxytriazolopyrimidine of the formula (IV) is generally reacted with an excess of halogenating agent. Work-up is carried out by customary methods.

Suitable diluents for carrying out the process (c) are all inert inorganic solvents customary for such reactions. Preference is given to using alcohols, such as methanol, ethanol, n-propanol, i-propanol, n-butanol and tert-butanol.

Suitable acid binders for carrying out the process (c) are all inorganic and organic bases customary for such reactions. Preference is given to using tertiary amines, such as tributylamine or pyridine. It is also possible for excess amine to act as diluent.

When carrying out the process (c), the temperatures can be varied within a relatively wide range. In general, the process is carried out at temperatures between 20°C and 200°C, preferably between 50°C and 180°C.

When carrying out the process (c), heteroarylmalonic esters of the formula (V) and aminotriazole of the formula (VI) are generally reacted in equivalent amounts. However, it is also possible to use an excess of one component or the other. Work-up is carried out by customary methods.

Suitable diluents for carrying out the processes (d) and (e) according to the invention are in each case all customary inert organic solvents. Preference is given to using halogenated hydrocarbons, such as, for example, chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride, dichloroethane or trichloroethane; ethers, such as diethyl ether, diisopropyl ether,

methyl t-butyl ether, methyl t-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole; nitriles, such as acetonitrile, propionitrile, n- or i-butyronitrile or benzonitrile; amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide; sulfoxides, such as dimethyl sulfoxide; sulfones, such as sulfolane; alcohols, such as methanol, ethanol, n- or i-propanol, n-, i-, sec- or tert-butanol, ethanediol, propane-1,2-diol, ethoxyethanol, methoxyethanol, diethylene glycol monomethyl ether, diethylene glycol monoethyl ether, mixtures thereof with water or else pure water.

Suitable copper salts for carrying out the processes (d) and (e) according to the invention are in each case customary copper salts. Preference is given to using copper(I) chloride or copper(I) bromide.

Suitable acid acceptors for carrying out the processes (d) and (e) according to the invention are in each case all customary inorganic or organic bases. Preference is given to using alkaline earth metal or alkali metal hydrides, hydroxides, amides, alkoxides, acetates, carbonates or bicarbonates, such as, for example, sodium hydride, sodium amide, lithium diisopropylamide, sodium methoxide, sodium ethoxide, potassium tert-butoxide, sodium hydroxide, potassium hydroxide, sodium acetate, potassium acetate, calcium acetate, sodium carbonate, potassium carbonate, potassium bicarbonate and sodium bicarbonate, and furthermore ammonium compounds, such as ammonium hydroxide, ammonium acetate and ammonium carbonate, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, N,N-dimethylbenzylamine, pyridine, N-methylpiperidine, N-methylmorpholine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

When carrying out the processes (d) and (e) according to the invention, the reaction temperatures can also be varied within a relatively wide range. In general, the processes are carried out at temperatures between 0°C and 150°C, preferably at temperatures between 0°C and 80°C.

When carrying out the process (d) according to the invention, in general from 1 to 15 mol, preferably from 1.3 to 8 mol, of malonic ester are employed per mole of halopyridine. Work-up is carried out by customary methods.

When carrying out the process (e) according to the invention, in general from 1 to 15 mol, preferably from 1.3 to 8 mol, of malonic ester are employed per mole of halopyrimidine. Work-up is again carried out by customary methods.

The processes according to the invention are generally carried out under atmospheric pressure. However, it is also possible to operate under elevated or reduced pressure.

The compounds according to the invention have potent microbicidal activity and can be employed for controlling unwanted microorganisms, such as fungi and bacteria, in crop protection and in the protection of materials.

Fungicides can be employed in crop protection for controlling Plasmodiophoromycetes, Oomycetes, Chytridiomycetes, Zygomycetes, Ascomycetes, Basidiomycetes and Deuteromycetes.

Bactericides can be employed in crop protection for controlling Pseudomonadaceae, Rhizobiaceae, Enterobacteriaceae, Corynebacteriaceae and Streptomycetaceae.

Some pathogens causing fungal and bacterial diseases which come under the generic names listed above may be mentioned as examples, but not by way of limitation:

Xanthomonas species, such as, for example, Xanthomonas campestris pv. oryzae;

Pseudomonas species, such as, for example, Pseudomonas syringae pv. lachrymans;

Erwinia species, such as, for example, Erwinia amylovora;

Pythium species, such as, for example, Pythium ultimum;

Phytophthora species, such as, for example, Phytophthora infestans;

Pseudoperonospora species, such as, for example, Pseudoperonospora humuli or Pseudoperonospora cubensis;

Plasmopara species, such as, for example, Plasmopara viticola;

Bremia species, such as, for example, Bremia lactucae;

Peronospora species, such as, for example, Peronospora pisi or P. brassicae;

Erysiphe species, such as, for example, Erysiphe graminis;

Sphaerotheca species, such as, for example, Sphaerotheca fuliginea;

Podosphaera species, such as, for example, Podosphaera leucotricha;

Venturia species, such as, for example, Venturia inaequalis;

Pyrenophora species, such as, for example, Pyrenophora teres or P. graminea (conidia form: Drechslera, syn: Helminthosporium);

Cochliobolus species, such as, for example, Cochliobolus sativus (conidia form: Drechslera, syn: Helminthosporium);

Uromyces species, such as, for example, Uromyces appendiculatus;

Puccinia species, such as, for example, Puccinia recondita;

Sclerotinia species, such as, for example, Sclerotinia sclerotiorum;

Tilletia species, such as, for example, Tilletia caries;

Ustilago species, such as, for example, Ustilago nuda or Ustilago avenae;

Pellicularia species, such as, for example, Pellicularia sasakii;

Pyricularia species, such as, for example, Pyricularia oryzae;

Fusarium species, such as, for example, Fusarium culmorum;

Botrytis species, such as, for example, Botrytis cinerea;

Septoria species, such as, for example, Septoria nodorum;

Leptosphaeria species, such as, for example, Leptosphaeria nodorum;

Cercospora species, such as, for example, Cercospora canescens;

Alternaria species, such as, for example, Alternaria brassicae; and

Pseudocercosporella species, such as, for example, Pseudocercosporella herpotrichoides.

The active compounds according to the invention also show a strong invigorating action in plants. Accordingly, they are suitable for mobilizing the internal defenses of the plant against attack by unwanted microorganisms.

In the present context, plant-invigorating (resistance-inducing) compounds are to be understood as meaning substances which are capable of stimulating the defense system of plants such that, when the treated plants are subsequently inoculated with unwanted microorganisms, they display substantial resistance to these microorganisms.

In the present case, unwanted microorganisms are to be understood as meaning phytopathogenic fungi, bacteria and viruses. The compounds according to the invention can thus be used to protect plants within a certain period of time after treatment against attack by the pathogens mentioned. The period of time for which this protection is achieved generally extends for 1 to 10 days, preferably 1 to 7 days, from the treatment of the plants with the active compounds.

The fact that the active compounds are well tolerated by plants at the concentrations required for controlling plant diseases permits the treatment of above-ground parts of plants, of propagation stock and seeds, and of the soil.

The active compounds according to the invention can be employed with particularly good results for controlling cereal diseases, such as, for example, against Erysiphe species, and of diseases in viticulture and in the cultivation of fruit and vegetables, such as, for example, against Botrytis, Venturia, Sphaerotheca and Podosphaera species.

The active compounds according to the invention are also suitable for increasing the yield of crops. In addition, they show reduced toxicity and are well tolerated by plants.

If appropriate, the active compounds according to the invention can, at certain concentrations and application rates, also be employed as herbicides, for regulating plant growth and for controlling animal pests. If appropriate, they can also be used as intermediates or precursors in the synthesis of other active compounds.

According to the invention, it is possible to treat all plants and parts of plants. Plants are to be understood here as meaning all plants and plant populations, such as desired and undesired wild plants or crop plants (including naturally occurring crop plants). Crop plants can be plants which can be obtained by conventional breeding and optimization methods or by biotechnological and genetic engineering methods or combinations of these methods, including the transgenic plants and including plant cultivars which can or cannot be protected by plant breeders' certificates. Parts of plants are to be understood as meaning all above-ground and below-ground parts and organs of

plants, such as shoot, leaf, flower and root, examples which may be mentioned being leaves, needles, stems, trunks, flowers, fruit-bodies, fruits and seeds and also roots, tubers and rhizomes. Parts of plants also include harvested material and vegetative and generative propagation material, for example seedlings, tubers, rhizomes, cuttings and seeds.

The treatment of the plants and parts of plants according to the invention with the active compounds is carried out directly or by action on their environment, habitat or storage area according to customary treatment methods, for example by dipping, spraying, evaporating, atomizing, broadcasting, brushing-on and, in the case of propagation material, in particular in the case of seeds, furthermore by one- or multilayer coating.

In the protection of materials, the compounds according to the invention can be employed for protecting industrial materials against infection with, and destruction by, unwanted microorganisms.

Industrial materials in the present context are understood as meaning non-living materials which have been prepared for use in industry. For example, industrial materials which are intended to be protected by active compounds according to the invention from microbial change or destruction can be tackifiers, sizes, paper and board, textiles, leather, wood, paints and plastic articles, cooling lubricants and other materials which can be infected with, or destroyed by, microorganisms. Parts of production plants, for example cooling-water circuits, which may be impaired by the proliferation of microorganisms may also be mentioned within the scope of the materials to be protected. Industrial materials which may be mentioned within the scope of the present invention are preferably adhesives, sizes, paper and board, leather, wood, paints, cooling lubricants and heat-transfer liquids, particularly preferably wood.

Microorganisms capable of degrading or changing the industrial materials which may be mentioned are, for example, bacteria, fungi, yeasts, algae and slime organisms. The active compounds according to the invention preferably act against fungi, in particular molds, wood-discoloring and wood-destroying fungi (Basidiomycetes) and against slime organisms and algae.

Microorganisms of the following genera may be mentioned as examples:

Alternaria, such as Alternaria tenuis,

Aspergillus, such as Aspergillus niger,

Chaetomium, such as Chaetomium globosum,

Coniophora, such as Coniophora puetana,

Lentinus, such as Lentinus tigrinus,

Penicillium, such as Penicillium glaucum,

Polyporus, such as Polyporus versicolor,

Aureobasidium, such as Aureobasidium pullulans,

Sclerophoma, such as Sclerophoma pityophila,

Trichoderma, such as Trichoderma viride,

Escherichia, such as Escherichia coli,

Pseudomonas, such as Pseudomonas aeruginosa, and

Staphylococcus, such as Staphylococcus aureus.

Depending on their particular physical and/or chemical properties, the active compounds can be converted into the customary formulations, such as solutions, emulsions, suspensions, powders, foams, pastes, granules, aerosols and microencapsulations in polymeric substances and in coating compositions for seeds, and ULV cool and warm fogging formulations.

These formulations are produced in a known manner, for example by mixing the active compounds with extenders, that is liquid solvents, liquefied gases under pressure, and/or solid carriers, optionally with the use of surfactants, that is emulsifiers and/or dispersants, and/or foam formers. If the extender used is water, it is also possible to employ, for example, organic solvents as auxiliary solvents. Essentially, suitable liquid solvents are: aromatics such as xylene, toluene or alkylnaphthalenes, chlorinated aromatics or chlorinated aliphatic hydrocarbons such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic hydrocarbons such as cyclohexane or paraffins, for example petroleum fractions, alcohols such as butanol or glycol and their ethers and esters, ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents such as dimethylformamide or dimethyl sulfoxide, or else water. Liquefied gaseous extenders or carriers are to be understood as meaning those liquids which are gaseous at standard temperature and under atmospheric pressure, for example aerosol

propellants such as halogenated hydrocarbons, or else butane, propane, nitrogen and carbon dioxide. Suitable solid carriers are: for example ground natural minerals such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals such as finely divided silica, alumina and silicates. Suitable solid carriers for granules are: for example crushed and fractionated natural rocks such as calcite, pumice, marble, sepiolite and dolomite, or else synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, corn cobs and tobacco stalks. Suitable emulsifiers and/or foam formers are: for example nonionic and anionic emulsifiers, such as polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulfonates, alkyl sulfates, arylsulfonates, or else protein hydrolyzates. Suitable dispersants are: for example lignosulfite waste liquors and methylcellulose.

Tackifiers such as carboxymethylcellulose, natural and synthetic polymers in the form of powders, granules or latices, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, or else natural phospholipids such as cephalins and lecithins and synthetic phospholipids can be used in the formulations. Other possible additives are mineral and vegetable oils.

It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs such as alizarin dyestuffs, azo dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

The formulations generally comprise between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.

The active compounds according to the invention can, as such or in their formulations, also be used in a mixture with known fungicides, bactericides, acaricides, nematicides or insecticides, to broaden, for example, the activity spectrum or to prevent development of resistance. In many cases, synergistic effects are obtained, i.e. the activity of the mixture is greater than the activity of the individual components.

Suitable mixing components are, for example, the following compounds:

Fungicides:

2-phenylphenol; 8-hydroxyquinoline sulfate; acibenzolar-S-methyl; aldimorph; amidoflumet; ampropylfos; ampropylfos-potassium; andoprim; anilazine; azaconazole; azoxystrobin; benalaxyl; benalaxyl-M, benodanil; benomyl; benthiavalicarb-isopropyl; benzamacril; benzamacril-isobutyl; bilanafos; binapacryl; biphenyl; bitertanol; blasticidin-S; boscalid; bromuconazole; bupirimate;

buthiobate; butylamine; calcium polysulfide; capsimycin; captafol; captan; carbendazim; carboxin; carpropamid; carvone; chinomethionat; chlobenthiazone; chlorfenazole; chloroneb; chlorothalonil; chlozolinate; clozylacon; cyazofamid; cyflufenamid; cymoxanil; cyproconazole; cyprodinil; cyprofuram; Dagger G; debacarb; dichlofluanid; dichlone; dichlorophen; diclocymet; diclomezine; dicloran; diethofencarb; difenoconazole; diflumetorim; dimethirimol; dimethomorph; dimoxystrobin; diniconazole; diniconazole-M; dinocap; diphenylamine; dipyrithione; ditalimfos; dithianon; dodine; drazoxolon; edifenphos; epoxiconazole; ethaboxam; ethirimol; etridiazole; famoxadone; fenamidone; fenapanil; fenarimol; fenbuconazole; fenfuram; fenhexamid; fenitropan; fenoxanil; fenpiclonil; fenpropidin; fenpropimorph; ferbam; fluazinam; flubenzimine; fludioxonil; flumetover; flumorph; fluoromide; fluoxastrobin; fluquinconazole; flurprimidol; flusilazole; flusulfamide; flutolanil; flutriafol; folpet; fosetyl-Al; fosetyl-sodium; fuberidazole; furalaxyl; furametpyr; furcarbanil; furmecyclox; guazatine; hexachlorobenzene; hexaconazole; hymexazole; imazalil; imibenconazole; iminoctadine triacetate; iminoctadine tris(albesilate); iodocarb; ipconazole; iprobenfos; iprodione; iprovalicarb; irumamycin; isoprothiolane; isovaledione; kasugamycin; kresoxim-methyl; mancozeb; maneb; meferimzone; mepanipyrim; mepronil; metalaxyl; metalaxyl-M; metconazole; methasulfocarb; methfuroxam; metiram; metominostrobin; metsulfovax; mildiomycin; myclobutanil; myclozolin; natamycin; nicobifen; nitrothal-isopropyl; noviflumuron; nuarimol; ofurace; orysastrobin; oxadixyl; oxolinic acid; oxpoconazole; oxycarboxin; oxyfenthiin; paclobutrazole; pefurazoate; penconazole; pencycuron; phosdiphen; phthalide; picoxystrobin; piperalin; polyoxins; polyoxorim; probenazole; prochloraz; procymidone; propamocarb; propanosine-sodium; propiconazole; propineb; proquinazid; prothioconazole; pyraclostrobin; pyrazophos; pyrifenox; pyrimethanil; pyroquilon; pyroxyfur; pyrrolenitrine; quinconazole; quinoxyfen; quintozene; simeconazole; spiroxamine; sulfur; tebuconazole; tecloftalam; tecnazene; tetcyclacis; tetraconazole; thiabendazole; thicyofen; thifluzamide; thiophanate-methyl; thiram; tioxymid; tolclofos-methyl; tolylfluanid; triadimefon; triadimenol; triazbutil; triazoxide; tricyclamide; tricyclazole; tridemorph; trifloxystrobin; triflumizole; triforine; triticonazole; uniconazole; validamycin A; vinclozolin; zineb; ziram; zoxamide; (2S)-N-[2-[4-[[3-(4-chlorophenyl)-2-propynyl]oxy]-3-methoxyphenyl]ethyl]-3-methyl-2-[(methylsulfonyl)amino]butanamide; 1-(1-naphthalenyl)-1H-pyrrole-2,5-dione; 2,3,5,6-tetrachloro-4-(methylsulfonyl)pyridine; 2-amino-4-methyl-N-phenyl-5-thiazolecarboxamide; 2-chloro-N-(2,3-dihydro-1,1,3-trimethyl-1H-inden-4-yl)-3-pyridinecarboxamide; 3,4,5-trichloro-2,6-pyridinedicarbonitrile; actinovate; cis-1-(4-chlorophenyl)-2-(1H-1,2,4-triazol-1-yl)cycloheptanol; methyl 1-(2,3-dihydro-2,2-dimethyl-1H-inden-1-yl)-1H-imidazole-5-carboxylate; monopotassium carbonate; N-(6-methoxy-3-pyridinyl)cyclopropanecarboxamide; N-butyl-8-(1,1-dimethylethyl)-1-oxaspiro[4.5]decane-3-amine; sodium tetracarbonate;

and copper salts and preparations, such as Bordeaux mixture; copper hydroxide, copper

naphthenate; copper oxychloride; copper sulfate; cufraneb; copper oxide; mancopper; oxine-copper.

Bactericides:

bronopol, dichlorophen, nitrapyrin, nickel dimethyldithiocarbamate, kasugamycin, octhilinone, furancarboxylic acid, oxytetracyclin, probenazole, streptomycin, tecloftalam, copper sulfate and other copper preparations.

Insecticides / acaricides / nematicides:

1. Acetylcholinesterase (AChE) inhibitors

1.1 carbamates (for example alanycarb, aldicarb, aldoxycarb, allyxycarb, aminocarb, azamethiphos, bendiocarb, benfuracarb, bufencarb, butacarb, butocarboxim, butoxycarboxim, carbaryl, carbofuran, carbosulfan, chloethocarb, coumaphos, cyanofenphos, cyanophos, dimetilan, ethiofencarb, fenobucarb, fenothiocarb, formetanate, furathiocarb, isoprocarb, metam-sodium, methiocarb, methomyl, metolcarb, oxamyl, pirimicarb, promecarb, propoxur, thiodicarb, thiofanox, triazamate, trimethacarb, XMC, xylylcarb)

1.2 organophosphates (for example acephate, azamethiphos, azinphos (-methyl, -ethyl), bromophos-ethyl, bromfenvinfos (-methyl), butathiofos, cadusafos, carbophenothion, chlorethoxyfos, chlorfenvinphos, chlormephos, chlorpyrifos (-methyl/-ethyl), coumaphos, cyanofenphos, cyanophos, chlorofenvinphos, demeton-S-methyl, demeton-S-methylsulfone, dialifos, diazinon, dichlofenthion, dichlorvos/DDVP, dicrotophos, dimethoate, dimethylvinphos, dioxabenzofos, disulfoton, EPN, ethion, ethoprophos, etrimfos, famphur, fenamiphos, fenitrothion, fensulfothion, fenthion, flupyrazofos, fonofos, formothion, fosmethilan, fosthiazate, heptenophos, iodofenphos, iprobenfos, isazofos, isofenphos, isopropyl o-salicylate, isoxathion, malathion, mecarbam, methacrifos, methamidophos, methidathion, mevinphos, monocrotophos, naled, omethoate, oxydemeton-methyl, parathion (-methyl/-ethyl), phenthoate, phorate, phosalone, phosmet, phosphamidon, phosphocarb, phoxim, pirimiphos (-methyl/-ethyl), profenofos, propaphos, propetamphos, prothiofos, prothoate, pyraclofos, pyridaphenthion, pyridathion, quinalphos, sebufos, sulfotep, sulprofos, tebupirimfos, temephos, terbufos, tetrachlorovinphos, thiometon, triazophos, triclorfon, vamidothion)

2. Sodium channel modulators/blockers of voltage-gated sodium channels

2.1 pyrethroids (for example acrinathrin, allethrin (d-cis-trans, d-trans), beta-cyfluthrin, bifenthrin, bioallethrin, bioallethrin-S-cyclopentyl-isomer, bioethanomethrin, biopermethrin, bioresmethrin, chlovaporthrin, cis-cypermethrin, cis-resmethrin, cis-permethrin, clocythrin, cycloprothrin, cyflu-

thrin, cyhalothrin, cypermethrin (alpha-, beta-, theta-, zeta-), cyphenothrin, DDT, deltamethrin, empenthrin (1R-isomer), esfenvalerate, etofenprox, fenfluthrin, fenpropathrin, fenpyrithrin, fenvalerate, flubrocythrinate, flucythrinate, flufenprox, flumethrin, fluvalinate, fubfenprox, gamma-cyhalothrin, imiprothrin, kadethrin, lambda-cyhalothrin, metofluthrin, permethrin (cis-, trans-), phenothrin (1R-trans isomer), prallethrin, profluthrin, protrifenbute, pyresmethrin, resmethrin, RU 15525, silafluofen, tau-fluvalinate, tefluthrin, terallethrin, tetramethrin (1R-isomer), tralomethrin, transfluthrin, ZXI 8901, pyrethrins (pyrethrum))

2.2 oxadiazines (for example indoxacarb)

3. Acetylcholine receptor agonists/antagonists

3.1 chloronicotinyls/neonicotinoids (for example acetamiprid, clothianidin, dinotefuran, imidacloprid, nitenpyram, nithiazine, thiacloprid, thiamethoxam)

3.2 nicotine, bensultap, cartap

4. Acetylcholine receptor modulators

4.1 spinosyns (for example spinosad)

5. Antagonists of GABA-gated chloride channels

5.1 cyclodiene organochlorines (for example camphechlor, chlordane, endosulfan, gamma-HCH, HCH, heptachlor, lindane, methoxychlor

5.2 fiproles (for example acetoprole, ethiprole, fipronil, vaniliprole)

6. Chloride channel activators

6.1 mectins (for example abamectin, avermectin, emamectin, emamectin-benzoate, ivermectin, milbemectin, milbemycin)

7. Juvenile hormone mimetics

(for example diofenolan, epofenonane, fenoxycarb, hydroprene, kinoprene, methoprene, pyriproxifen, triprene)

8. Ecdyson agonists/disruptors

8.1 diacylhydrazines (for example chromafenozide, halofenozide, methoxyfenozide, tebufenozide)

9. Chitin biosynthesis inhibitors

9.1 benzoylureas (for example bistrifluron, chlofluazuron, diflubenzuron, fluazuron, flucycloxuron, flufenoxuron, hexaflumuron, lufenuron, novaluron, noviflumuron, penfluron, teflubenzuron, triflumuron)

9.2 buprofezin

9.3 cyromazine

10. Inhibitors of oxidative phosphorylation, ATP disruptors

10.1 diafenthiuron

10.2 organotins (for example azocyclotin, cyhexatin, fenbutatin-oxide)

11. Decouplers of oxidative phosphorylation acting by interrupting the H-proton gradient

11.1 pyrroles (for example chlorfenapyr)

11.2 dinitrophenols (for example binapacryl, dinobuton, dinocap, DNOC)

12. Site-I electron transport inhibitors

12.1 METIs (for example fenazaquin, fenpyroximate, pyrimidifen, pyridaben, tebufenpyrad, tolfenpyrad)

12.2 hydramethylnone

12.3 dicofol

13. Site-II electron transport inhibitors

13.1 rotenone

14. Site-III electron transport inhibitors

14.1 acequinocyl, fluacrypyrim

15. Microbial disruptors of the insect gut membrane

Bacillus thuringiensis strains

16. Inhibitors of fat synthesis

16.1 tetronic acids (for example spirodiclofen, spiromesifen)

16.2 tetramic acids [for example 3-(2,5-dimethylphenyl)-8-methoxy-2-oxo-1-azaspiro[4.5]dec-3-en-4-yl ethyl carbonate (alias: carbonic acid, 3-(2,5-dimethylphenyl)-8-methoxy-2-oxo-1-azaspiro[4.5]dec-3-en-4-yl ethyl ester, CAS Reg. No.: 382608-10-8) and carbonic acid, cis-3-(2,5-dimethylphenyl)-8-methoxy-2-oxo-1-azaspiro[4.5]dec-3-en-4-yl ethyl ester (CAS Reg. No.: 203313-25-1)]

17. Carboxamides

(for example flonicamid)

18. Octopaminergic agonists

(for example amitraz)

19. Inhibitors of magnesium-stimulated ATPase

(for example propargite)

20. Phthalamides

(for example N^2-[1,1-dimethyl-2-(methylsulfonyl)ethyl]-3-iodo-N^1-[2-methyl-4-[1,2,2,2-tetrafluoro-1-(trifluoromethyl)ethyl]phenyl]-1,2-benzenedicarboxamide (CAS Reg. No.: 272451-65-7), flubendiamide)

21. Nereistoxin analogues

(for example thiocyclam hydrogen oxalate, thiosultap-sodium)

22. Biologicals, hormones or pheromones

(for example azadirachtin, Bacillus spec., Beauveria spec., codlemone, Metarrhizium spec., Paecilomyces spec., thuringiensin, Verticillium spec.)

23. Active compounds with unknown or unspecific mechanisms of action

23.1 fumigants (for example aluminum phosphide, methyl bromide, sulfuryl fluoride)

23.2 selective antifeedants (for example cryolite, flonicamid, pymetrozine)

23.3 mite growth inhibitors (for example clofentezine, etoxazole, hexythiazox)

23.4 amidoflumet, benclothiaz, benzoximate, bifenazate, bromopropylate, buprofezin, chinomethionat, chlorodimeform, chlorobenzilate, chloropicrin, clothiazoben, cycloprene, cyflumetofen, di-

cyclanil, fenoxacrim, fentrifanil, flubenzimine, flufenerim, flutenzin, gossyplure, hydramethylnone, japonilure, metoxadiazone, petroleum, piperonyl butoxide, potassium oleate, pyrafluprole, pyridalyl, pyriprole, sulfluramid, tetradifon, tetrasul, triarathene, verbutin,

furthermore the compound 3-methylphenyl propylcarbamate (Tsumacide Z), the compound 3-(5-chloro-3-pyridinyl)-8-(2,2,2-trifluoroethyl)-8-azabicyclo[3.2.1]octane-3-carbonitrile (CAS Reg. No. 185982-80-3) and the corresponding 3-endo-isomer (CAS Reg. No. 185984-60-5) (cf. WO 96/37494, WO 98/25923), and preparations which comprise insecticidally active plant extracts, nematodes, fungi or viruses.

A mixture with other known active compounds, such as herbicides, or with fertilizers and growth regulators, safeners and/or semiochemicals is also possible.

In addition, the compounds of the formula (I) according to the invention also have very good antimycotic activity. They have a very broad antimycotic activity spectrum in particular against dermatophytes and yeasts, molds and diphasic fungi (for example against Candida species such as Candida albicans, Candida glabrata) and Epidermophyton floccosum, Aspergillus species such as Aspergillus niger and Aspergillus fumigatus, Trichophyton species such as Trichophyton mentagrophytes, Microsporon species such as Microsporon canis and audouinii. The list of these fungi does by no means limit the mycotic spectrum which can be covered, but is only for illustration.

The active compounds can be used as such, in the form of their formulations or the use forms prepared therefrom, such as ready-to-use solutions, suspensions, wettable powders, pastes, soluble powders, dusts and granules. Application is carried out in a customary manner, for example by watering, spraying, atomizing, broadcasting, dusting, foaming, spreading, etc. It is furthermore possible to apply the active compounds by the ultra-low-volume method, or to inject the active compound preparation or the active compound itself into the soil. It is also possible to treat the seeds of the plants.

When using the active compounds according to the invention as fungicides, the application rates can be varied within a relatively wide range, depending on the kind of application. For the treatment of parts of plants, the active compound application rates are generally between 0.1 and 10 000 g/ha, preferably between 10 and 1000 g/ha. For seed dressing, the active compound application rates are generally between 0.001 and 50 g per kilogram of seed, preferably between 0.01 and 10 g per kilogram of seed. For the treatment of the soil, the active compound application rates are generally between 0.1 and 10 000 g/ha, preferably between 1 and 5 000 g/ha.

As already mentioned above, it is possible to treat all plants and their parts according to the invention. In a preferred embodiment, wild plant species and plant cultivars, or those obtained by conventional biological breeding, such as crossing or protoplast fusion, and parts thereof, are treated. In a further preferred embodiment, transgenic plants and plant cultivars obtained by genetic engineering, if appropriate in combination with conventional methods (Genetically Modified Organisms), and parts thereof, are treated. The term "parts" or "parts of plants" or "plant parts" has been explained above.

Particularly preferably, plants of the plant cultivars which are in each case commercially available or in use are treated according to the invention. Plant cultivars are to be understood as meaning plants having new properties ("traits") and which have been obtained by conventional breeding, by mutagenesis or by recombinant DNA techniques. They can be cultivars, varieties, bio- or genotypes.

Depending on the plant species or plant cultivars, their location and growth conditions (soils, climate, vegetation period, diet), the treatment according to the invention may also result in superadditive ("synergistic") effects. Thus, for example, reduced application rates and/or a widening of the activity spectrum and/or an increase in the activity of the substances and compositions which can be used according to the invention, better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to water or soil salt content, increased flowering performance, easier harvesting, accelerated maturation, higher harvest yields, better quality and/or a higher nutritional value of the harvested products, better storage stability and/or processability of the harvested products are possible which exceed the effects which were actually to be expected.

The transgenic plants or plant cultivars (i.e. those obtained by genetic engineering) which are preferably to be treated according to the invention include all plants which, in the genetic modification, received genetic material which imparted particularly advantageous useful properties ("traits") to these plants. Examples of such properties are better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to water or soil salt content, increased flowering performance, easier harvesting, accelerated maturation, higher harvest yields, better quality and/or a higher nutritional value of the harvested products, better storage stability and/or processability of the harvested products. Further and particularly emphasized examples of such properties are a better defense of the plants against animal and microbial pests, such as against insects, mites, phytopathogenic fungi, bacteria and/or viruses, and also increased tolerance of the plants to certain herbicidally active compounds. Examples of transgenic plants which may be mentioned are the important crop plants, such as cereals (wheat, rice), corn, soy beans, potatoes, cotton, tobacco, oilseed rape and also fruit plants (with the fruits apples, pears, citrus fruits and

grapes), and particular emphasis is given to corn, soy beans, potatoes, cotton, tobacco and oilseed rape. Traits that are particularly emphasized are increased defense of the plants against insects, arachnids, nematodes and slugs and snails by toxins formed in the plants, in particular those formed in the plants by the genetic material from Bacillus thuringiensis (for example by the genes CryIA(a), CryIA(b), CryIA(c), CryIIA, CryIIIA, CryIIIB2, Cry9c Cry2Ab, Cry3Bb and CryIF and also combinations thereof) (hereinbelow referred to as "Bt plants"). Traits that are also particularly emphasized are the increased defense of the plants against fungi, bacteria and viruses by systemic acquired resistance (SAR), systemin, phytoalexins, elicitors and resistance genes and correspondingly expressed proteins and toxins. Traits that are furthermore particularly emphasized are the increased tolerance of the plants to certain herbicidally active compounds, for example imidazolinones, sulfonylureas, glyphosate or phosphinotricin (for example the "PAT" gene). The genes which impart the desired traits in question can also be present in combination with one another in the transgenic plants. Examples of "Bt plants" which may be mentioned are corn varieties, cotton varieties, soy bean varieties and potato varieties which are sold under the trade names YIELD GARD® (for example corn, cotton, soy beans), KnockOut® (for example corn), StarLink® (for example corn), Bollgard® (cotton), Nucoton® (cotton) and NewLeaf® (potato). Examples of herbicide-tolerant plants which may be mentioned are corn varieties, cotton varieties and soy bean varieties which are sold under the trade names Roundup Ready® (tolerance to glyphosate, for example corn, cotton, soy bean), Liberty Link® (tolerance to phosphinotricin, for example oilseed rape), IMI® (tolerance to imidazolinones) and STS® (tolerance to sulfonylureas, for example corn). Herbicide-resistant plants (plants bred in a conventional manner for herbicide tolerance) which may be mentioned also include the varieties sold under the name Clearfield® (for example corn). Of course, these statements also apply to plant cultivars which have these genetic traits or genetic traits still to be developed, and which will be developed and/or marketed in the future.

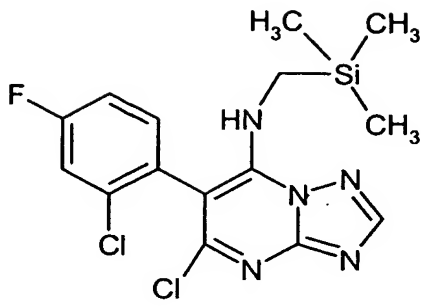
The plants listed can be treated according to the invention in a particularly advantageous manner with the compounds of the general formula (I) or the active compound mixtures according to the invention. The preferred ranges stated above for the active compounds or mixtures also apply to the treatment of these plants. Particular emphasis is given to the treatment of plants with the compounds or mixtures specifically mentioned in the present text.

The compounds of the formula (I) according to the invention are furthermore suitable for suppressing the growth of tumor cells in humans and mammals. This is based on an interaction of the compounds according to the invention with tubulin and microtubuli and by promoting microtubuli polymerization.

For this purpose, it is possible to administer an effective amount of one or more compounds of the formula (I) or pharmaceutically acceptable salts thereof.

The preparation and the use of the active compounds according to the invention is illustrated in the examples below.

Example 1



At room temperature, 0.143 g (1.386 mmol) of trimethylsilylmethylamine and 0.216 g (1.890 mmol) of K_2CO_3 were added to a solution of 0.400 g (1.260 mmol) of 5,7-dichloro-6-(2-chloro-4-fluorophenyl)-[1,2,4]triazolo[1,5-a]pyrimidine in 10 ml of acetonitrile, and the mixture was then stirred overnight. The mixture was then stirred into hydrochloric acid, and the precipitated product was washed with water and dried. This gave 0.31 g (58% of theory) of product.

HPLC: logP = 3.61

Example 2

a) Preparation of (1-iodoethyl)trimethylsilane

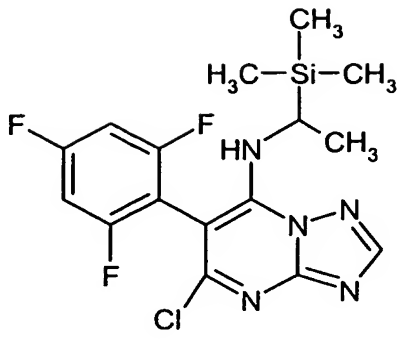
2.4 g (17.557 mmol) of (1-chloroethyl)trimethylsilane were stirred into 15 ml of acetone, 6.579 g (43.893 mmol) of sodium iodide were added and the mixture was boiled at reflux for 1 day. The mixture was taken up in water and ether and the aqueous phase was extracted twice with ether. The organic phase was dried and the solvent was removed under reduced pressure.

This gave 3.8 g (95 % of theory) of product.

b) (1-Aminoethyl)trimethylsilane

In an autoclave, 3.8 g (16.656 mmol) (1-iodomethyl)trimethylsilane were condensed with 47.670 g (2799.014 mmol) of ammonia, and the mixture was stirred at 135°C and a pressure of 91 bar for 2.5 hours. The reaction mixture was stirred into CH_2Cl_2, the solvent was removed, 6n aqueous sodium hydroxide solution was added and the mixture was extracted with ether. Removal of the ester under reduced pressure gave 1.4 g (70% of theory) of the product.

c)

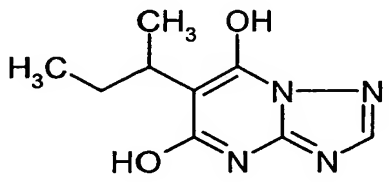


At room temperature, 0.217 g (1.567 mmol) of K_2CO_3 was added to a solution of 0.500 g (1.567 mmol) of 5,7-dichloro-6-(2,4,6-trifluorophenyl)-[1,2,4]triazolo[1,5-a]pyrimidine in 10 ml of acetonitrile and 0.202 g (1.724 mmol) of 1-(trimethylsilyl)ethylamine and the mixture was stirred at room temperature overnight and then at 60°C for another 6 hours. The solvent was removed under reduced pressure and the residue was taken up in CH_2Cl_3 and 1N hydrochloric acid. Drying and concentration of the organic phase was followed by chromatographic purification (petroleum ether/methyl isobutyl ketone 3:1) of the residue that remained. This gave 0.35 g (51% of theory) of product.

HPLC: logP = 3.71

Example 25

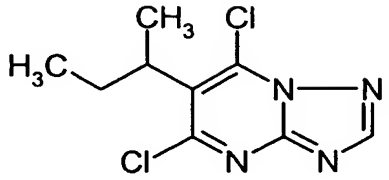
a)



2.6 g (0.012 mol) of diethyl sec-butylmalonate were initially charged in 9.5 g of dimethylformamide. At room temperature, 1 g (0.012 mol) of 3-amino-1,2,4-triazole and 1.8 g (0.013 mol) of 1.8-diazabicyclo(5.4.0)undec-7-ene were added. The mixture was stirred at 100°C for 6 hours. The mixture was then poured into water and extracted with chloroform. The aqueous phase was concentrated and the residue that remained was stirred with concentrated hydrochloric acid, filtered off with suction and dried.

This gave 0.5 g of 6-sec-butyl-7-hydroxy[1,2,4]triazolo[1,5-a]pyrimidin-5(4H)-one (log p = 0.70; HPCL content: 96.2%).

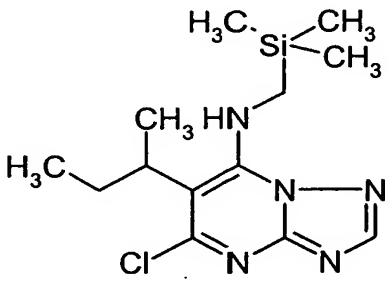
b)



2.6 g (0.093 mol) of diethyl sec-butylmalonate were mixed with 7.8 g (0.093 mol) of 3-amino-1,2,4-triazole and 19 g (0.102 mol) of tri-n-butylamine, and the mixture was stirred at 180°C for 6 hours, with distillative removal of the ethanol formed during the reaction. Volatile components were then distilled off at 10 Torr. The residue was reacted further without purification.

The crude 6-sec-butyl-7-hydroxy[1,2,4]triazolo[1,5-a]pyrimidin-5(4H)-one was dissolved in 8.9 g (0.827 mol) of phosphoryl chloride, and 11 g (0.053 mol) of phosphorus pentachloride were added at room temperature. The reaction mixture was heated at 110°C for 3 hours. Volatile components were then removed at 10 Torr, the residue was taken up in dichloromethane and the solution was washed with ice-water. After drying and distillative removal of the solvent, the residue was chromatographed on silica gel in a mixture of petroleum ether : tert-butyl methyl ether = 2 : 1. This gave 3.5 g of 5,7-dichloro-6-sec-butyl[1,2,4]triazolo[1,5-a]pyrimidine (log p = 2.40; HPLC content: 96.4 %).

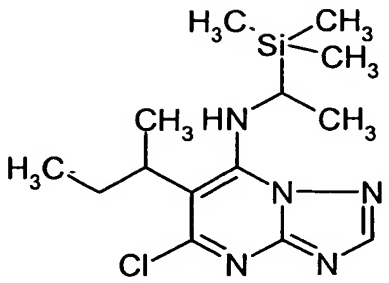
c)



0.5 g (0.002 mol) of 5,7-dichloro-6-sec-butyl[1,2,4]triazolo[1,5-a]pyrimidine was initially charged in 7.8 g of acetonitrile. 0.7 g (0.005 mol) of potassium carbonate and 0.31 g (0.002 mol) of 2-trimethylsilyl-1-aminoethane were added. The reaction mixture was stirred at room temperature for 16 hours. The mixture was then acidified with hydrochloric acid and extracted with diethyl ether. The organic phase was dried and concentrated. The residue was stirred with diethyl ether and then chromatographed on silica gel in a mixture of cychlohexane : ethyl acetate = 8 : 2.

This gave 0.3 g of 5-chloro-6-sec-butyl-7-(1-trimethylsilylethylamino)[1,2,4]triazolo[1,5-a]pyrimidine (log p = 3.87; HPLC content: 93%).

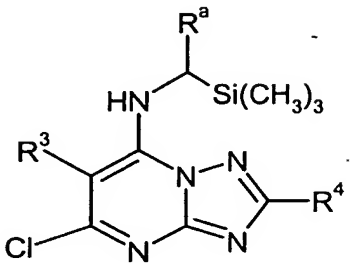
Example 26



0.5 g (0.002 mol) of 5,7-dichloro-6-sec-butyl[1,2,4]triazolo[1,5-a]pyrimidine was initially charged in 7.8 g of acetonitrile. 0.42 g (0.003 mol) of potassium carbonate and 0.21 g (0.002 mol) of trimethylsilylmethylamine were added. The reaction mixture was stirred at room temperature for 16 hours. The mixture was then acidified with hydrochloric acid and extracted with diethyl ether. The organic phase was dried and concentrated. The residue was stirred with diethyl ether and then filtered off with suction. This gave 0.24 g of 5-chloro-6-sec-butyl-7-trimethyl-silylmethylamino[1,2,4]triazolo[1,5-a]pyrimidine (log p = 3.34; HPLC content: 100%).

The compounds of the formula (I) listed in tables 1 to 5 below are or were also obtained analogously to the methods described above.

Table 1

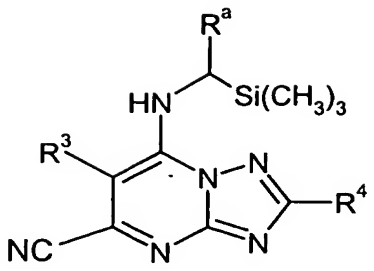


Ex. No.	R^a	R^3	R^4	logP
1	H	2-Cl-4-F-phenyl	H	3.61
2	CH_3	2,4,6-trifluorophenyl	H	3.71
3	H	2-Cl-6-F-phenyl	H	3.43
4	H	2,4,6-trifluorophenyl	H	3.39
5	CH_3	2-Cl-6-F-phenyl	H	3.79
6	CH_3	2-Cl-4-F-phenyl	H	3.95
7	CH_3	2-Cl-phenyl	H	3.83
8	H	3-Cl-5-(CF_3)-pyridin-2-yl	i-propyl	4.51
9	CH_3	5-F-pyrimidin-4-yl	i-propyl	3.61
10	CH_3	3-(CF_3)-pyridin-2-yl	H	3.06
11	CH_3	2-Cl-6-F-phenyl	Cyclopropyl	4.65
12	H	2-Cl-6-F-phenyl	Cyclopropyl	4.22
13	H	2,5-difluorophenyl	Cyclopropyl	3.99
14	CH_3	2,5-difluorophenyl	Cyclopropyl	4.35
15	CH_3	2,5-difluorophenyl	i-propyl	4.58
16	H	2,5-difluorophenyl	i-propyl	4.21
17	CH_3	2,5-difluorophenyl	Methyl	3.73
18	H	2,5-difluorophenyl	Methyl	3.40
19	CH_3	5-F-pyrimidin-4-yl	Cyclopropyl	2.99
20	H	2-Cl-phenyl	Cyclopropyl	4.23
21	CH_3	2-Cl-phenyl	Cyclopropyl	4.66
22	H	5-F-pyrimidin-4-yl	Cyclopropyl	3.02

Ex. No.	R^a	R^3	R^4	logP
23	CH_3	5-Cl-pyrimidin-4-yl	H	2.70
24	H	5-Cl-pyrimidin-4-yl	H	2.44
25	CH_3	sec-butyl	H	3.87
26	H	sec-butyl	H	3.34
27	H	5-F-pyrimidin-4-yl	Methyl	2.43
28	CH_3	5-F-pyrimidin-4-yl	Methyl	2.73
29	H	N(-CH_2-CH_2-CH_2-CH_2-CH_2-)	H	
30	CH_3	N(-CH_2-CH_2-CH_2-CH_2-CH_2-)	H	
31	H	N(-$CHCH_3$-CH_2-CH_2-CH_2-CH_2-)	H	
32	H	N(-$CHCH_3$-CH_2-CH_2-CH_2-CH_2-)	H	
33	H	N(-$CHCH_3$-CH_2-O-CH_2-CH_2-)	H	
34	CH_3	N(-$CHCH_3$-CH_2-O-CH_2-CH_2-)	H	
35	H	3,5-dimethylpyrazol-1-yl	H	
36	CH_3	3,5-dimethylpyrazol-1-yl	H	
37	H	N(CH_3)(C_2H_5)	H	
38	CH_3	N(CH_3)(C_2H_5)	H	
39	H	2,5-difluorophenyl	H	3.26
40	CH_3	2,5-difluorophenyl	H	3.57
41	H	5-F-pyrimidin-4-yl	H	2.28
42	CH_3	5-F-pyrimidn-4-yl	H	2.57

The logP values were determined in accordance with EEC Directive 79/831 Annex V. A8 by HPLC (gradient method, acetonitrile/0.1% aqueous phosphoric acid.

Table 2

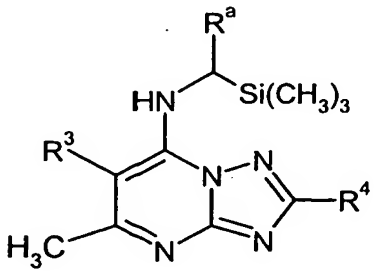


Ex. No.	R^a	R^3	R^4	logP
43	H	2-Cl-4-F-phenyl	H	
44	CH_3	2,4,6-trifluorophenyl	H	
45	H	2-Cl-6-F-phenyl	H	
46	H	2,4,6-trifluorophenyl	H	
47	CH_3	2-Cl-6-F-phenyl	H	
48	CH_3	2-Cl-4-F-phenyl	H	
49	CH_3	2-Cl-phenyl	H	
50	H	3-Cl-5-(CF_3)-pyridin-2-yl	i-propyl	
51	CH_3	5-F-pyrimidin-4-yl	i-propyl	
52	CH_3	3-(CF_3)-pyridin-2-yl	H	
53	CH_3	2-Cl-6-F-phenyl	Cyclopropyl	
54	H	2-Cl-6-F-phenyl	Cyclopropyl	
55	H	2,5-difluorophenyl	Cyclopropyl	
56	CH_3	2,5-difluorophenyl	Cyclopropyl	
57	CH_3	2,5-difluorophenyl	i-propyl	
58	H	2,5-difluorophenyl	i-propyl	
59	CH_3	2,5-difluorophenyl	Methyl	
60	H	2,5-difluorophenyl	Methyl	
61	CH_3	5-F-pyrimidin-4-yl	Cyclopropyl	
62	H	2-Cl-phenyl	Cyclopropyl	
63	CH_3	2-Cl-phenyl	Cyclopropyl	
64	H	5-F-pyrimidin-4-yl	Cyclopropyl	
65	CH_3	5-Cl-pyrimidin-4-yl	H	
66	H	5-Cl-pyrimidin-4-yl	H	

Ex. No.	R^a	R^3	R^4	logP
67	CH_3	sec-butyl	H	
68	H	sec-butyl	H	
69	H	5-F-pyrimidin-4-yl	Methyl	
70	CH_3	5-F-pyrimidin-4-yl	Methyl	
71	H	N(-CH_2-CH_2-CH_2-CH_2-CH_2-)	H	
72	CH_3	N(-CH_2-CH_2-CH_2-CH_2-CH_2-)	H	
73	H	N(-$CHCH_3$-CH_2-CH_2-CH_2-CH_2-)	H	
74	H	N(-$CHCH_3$-CH_2-CH_2-CH_2-CH_2-)	H	
75	H	N(-$CHCH_3$-CH_2-O-CH_2-CH_2-)	H	
76	CH_3	N(-$CHCH_3$-CH_2-O-CH_2-CH_2-)	H	
77	H	3,5-dimethylpyrazol-1-yl	H	
78	CH_3	3,5-dimethylpyrazol-1-yl	H	
79	H	N(CH_3)(C_2H_5)	H	
80	CH_3	N(CH_3)(C_2H_5)	H	
81	H	2,5-difluorophenyl	H	
82	CH_3	2,5-difluorophenyl	H	
83	H	5-F-pyrimidin-4-yl	H	
84	CH_3	5-F-pyrimidin-4-yl	H	

The logP values were determined in accordance with EEC Directive 79/831 Annex V. A8 by HPLC (gradient method, acetonitrile/0.1% aqueous phosphoric acid.

Table 3

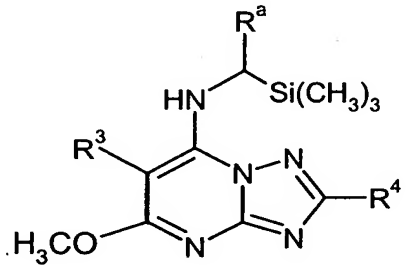


Ex. No.	R^a	R^3	R^4	logP
85	H	2-Cl-4-F-phenyl	H	
86	CH_3	2,4,6-trifluorophenyl	H	
87	H	2-Cl-6-F-phenyl	H	
88	H	2,4,6-trifluorophenyl	H	
89	CH_3	2-Cl-6-F-phenyl	H	
90	CH_3	2-Cl-4-F-phenyl	H	
91	CH_3	2-Cl-phenyl	H	
92	H	3-Cl-5-(CF_3)-pyridin-2-yl	i-propyl	
93	CH_3	5-F-pyrimidin-4-yl	i-propyl	
94	CH_3	3-(CF_3)-pyridin-2-yl	H	
95	CH_3	2-Cl-6-F-phenyl	Cyclopropyl	
96	H	2-Cl-6-F-phenyl	Cyclopropyl	
97	H	2,5-difluorophenyl	Cyclopropyl	
98	CH_3	2,5-difluorophenyl	Cyclopropyl	
99	CH_3	2,5-difluorophenyl	i-propyl	
100	H	2,5-difluorophenyl	i-propyl	
101	CH_3	2,5-difluorophenyl	Methyl	
102	H	2,5-difluorophenyl	Methyl	
103	CH_3	5-F-pyrimidin-4-yl	Cyclopropyl	
104	H	2-Cl-phenyl	Cyclopropyl	
105	CH_3	2-Cl-phenyl	Cyclopropyl	
106	H	5-F-pyrimidin-4-yl	Cyclopropyl	
107	CH_3	5-Cl-pyrimidin-4-yl	H	
108	H	5-Cl-pyrimidin-4-yl	H	
109	CH_3	sec-butyl	H	
110	H	sec-butyl	H	
111	H	5-F-pyrimidin-4-yl	Methyl	
112	CH_3	5-F-pyrimidin-4-yl	Methyl	
113	H	N(-CH_2-CH_2-CH_2-CH_2-CH_2-)	H	
114	CH_3	N(-CH_2-CH_2-CH_2-CH_2-CH_2-)	H	
115	H	N(-$CHCH_3$-CH_2-CH_2-CH_2-CH_2-)	H	
116	H	N(-$CHCH_3$-CH_2-CH_2-CH_2-CH_2-)	H	
117	H	N(-$CHCH_3$-CH_2-O-CH_2-CH_2-)	H	
118	CH_3	N(-$CHCH_3$-CH_2-O-CH_2-CH_2-)	H	

Ex. No.	R^a	R^3	R^4	logP
119	H	3,5-dimethylpyrazol-1-yl	H	
120	CH_3	3,5-dimethylpyrazol-1-yl	H	
121	H	$N(CH_3)(C_2H_5)$	H	
122	CH_3	$N(CH_3)(C_2H_5)$	H	
123	H	2,5-difluorophenyl	H	
124	CH_3	2,5-difluorophenyl	H	
125	H	5-F-pyrimidin-4-yl	H	
126	CH_3	5-F-pyrimidin-4-yl	H	

The logP values were determined in accordance with EEC Directive 79/831 Annex V. A8 by HPLC (gradient method, acetonitrile/0.1% aqueous phosphoric acid.

Table 4

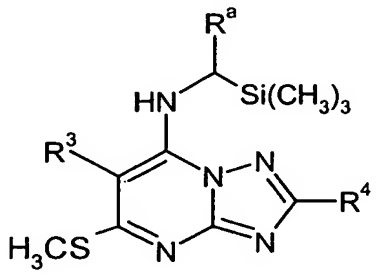


Ex. No.	R^a	R^3	R^4	logP
127	H	2-Cl-4-F-phenyl	H	
128	CH_3	2,4,6-trifluorophenyl	H	
129	H	2-Cl-6-F-phenyl	H	
130	H	2,4,6-trifluorophenyl	H	
131	CH_3	2-Cl-6-F-phenyl	H	
132	CH_3	2-Cl-4-F-phenyl	H	
133	CH_3	2-Cl-phenyl	H	
134	H	3-Cl-5-(CF_3)-pyridin-2-yl	i-propyl	
135	CH_3	5-F-pyrimidin-4-yl	i-propyl	

Ex. No.	R^a	R^3	R^4	logP
136	CH_3	3-(CF_3)-pyridin-2-yl	H	
137	CH_3	2-Cl-6-F-phenyl	Cyclopropyl	
138	H	2-Cl-6-F-phenyl	Cyclopropyl	
139	H	2,5-difluorophenyl	Cyclopropyl	
140	CH_3	2,5-difluorophenyl	Cyclopropyl	
141	CH_3	2,5-difluorophenyl	i-Propyl	
142	H	2,5-difluorophenyl	i-Propyl	
143	CH_3	2,5-difluorophenyl	Methyl	
144	H	2,5-difluorophenyl	Methyl	
145	CH_3	5-F-pyrimidin-4-yl	Cyclopropyl	
146	H	2-Cl-phenyl	Cyclopropyl	
147	CH_3	2-Cl-phenyl	Cyclopropyl	
148	H	5-F-pyrimidin-4-yl	Cyclopropyl	
149	CH_3	5-Cl-pyrimidin-4-yl	H	
150	H	5-Cl-pyrimidin-4-yl	H	
151	CH_3	sec-butyl	H	
152	H	sec-butyl	H	
153	H	5-F-pyrimidin-4-yl	Methyl	
154	CH_3	5-F-pyrimidin-4-yl	Methyl	
155	H	N(-CH_2-CH_2-CH_2-CH_2-CH_2-)	H	
156	CH_3	N(-CH_2-CH_2-CH_2-CH_2-CH_2-)	H	
157	H	N(-$CHCH_3$-CH_2-CH_2-CH_2-CH_2-)	H	
158	H	N(-$CHCH_3$-CH_2-CH_2-CH_2-CH_2-)	H	
159	H	N(-$CHCH_3$-CH_2-O-CH_2-CH_2-)	H	
160	CH_3	N(-$CHCH_3$-CH_2-O-CH_2-CH_2-)	H	
161	H	3,5-dimethylpyrazol-1-yl	H	
162	CH_3	3,5-dimethylpyrazol-1-yl	H	
163	H	N(CH_3)(C_2H_5)	H	
164	CH_3	N(CH_3)(C_2H_5)	H	
165	H	2,5-difluorophenyl	H	
166	CH_3	2,5-difluorophenyl	H	
167	H	5-F-pyrimidin-4-yl	H	
168	CH_3	5-F-pyrimidin-4-yl	H	

The logP values were determined in accordance with EEC Directive 79/831 Annex V. A8 by HPLC (gradient method: acetonitrile/0.1% aqueous phosphoric acid).

Table 5



Ex. No.	R^a	R^3	R^4	logP
169	H	2-Cl-4-F-phenyl	H	
170	CH_3	2,4,6-trifluorophenyl	H	
171	H	2-Cl-6-F-phenyl	H	
172	H	2,4,6-trifluorophenyl	H	
173	CH_3	2-Cl-6-F-phenyl	H	
174	CH_3	2-Cl-4-F-phenyl	H	
175	CH_3	2-Cl-phenyl	H	
176	H	3-Cl-5-(CF_3)-pyridin-2-yl	i-propyl	
177	CH_3	5-F-pyrimidin-4-yl	i-propyl	
178	CH_3	3-(CF_3)-pyridin-2-yl	H	
179	CH_3	2-Cl-6-F-phenyl	Cyclopropyl	
180	H	2-Cl-6-F-phenyl	Cyclopropyl	
181	H	2,5-difluorophenyl	Cyclopropyl	
182	CH_3	2,5-difluorophenyl	Cyclopropyl	
183	CH_3	2,5-difluorophenyl	i-propyl	
184	H	2,5-difluorophenyl	i-propyl	
185	CH_3	2,5-difluorophenyl	Methyl	
186	H	2,5-difluorophenyl	Methyl	
187	CH_3	5-F-pyrimidin-4-yl	Cyclopropyl	
188	H	2-Cl-phenyl	Cyclopropyl	

Ex. No.	R^a	R^3	R^4	logP
189	CH_3	2-Cl-phenyl	Cyclopropyl	
190	H	5-F-pyrimidin-4-yl	Cyclopropyl	
191	CH_3	5-Cl-pyrimidin-4-yl	H	
192	H	5-Cl-pyrimidin-4-yl	H	
193	CH_3	sec-butyl	H	
194	H	sec-butyl	H	
195	H	5-F-pyrimidin-4-yl	Methyl	
196	CH_3	5-F-pyrimidin-4-yl	Methyl	
197	H	N(-CH_2-CH_2-CH_2-CH_2-CH_2-)	H	
198	CH_3	N(-CH_2-CH_2-CH_2-CH_2-CH_2-)	H	
199	H	N(-$CHCH_3$-CH_2-CH_2-CH_2-CH_2-)	H	
200	H	N(-$CHCH_3$-CH_2-CH_2-CH_2-CH_2-)	H	
201	H	N(-$CHCH_3$-CH_2-O-CH_2-CH_2-)	H	
202	CH_3	N(-$CHCH_3$-CH_2-O-CH_2-CH_2-)	H	
203	H	3,5-dimethylpyrazol-1-yl	H	
204	CH_3	3,5-dimethylpyrazol-1-yl	H	
205	H	N(CH_3)(C_2H_5)	H	
206	CH_3	N(CH_3)(C_2H_5)	H	
207	H	2,5-difluorophenyl	H	
208	CH_3	2,5-difluorophenyl	H	
209	H	5-F-pyrimidin-4-yl	H	
210	CH_3	5-F-pyrimidin-4-yl	H	

The logP values were determined in accordance with EEC Directive 79/831 Annex V. A8 by HPLC (gradient method: acetonitrile/0.1% aqueous phosphoric acid).

Use examples

Example A

Podosphaera-test (Apple)/protective

Solvents: 24.5 parts by weight of acetone

24.5 parts by weight of dimethylacetamide

Emulsifier : 1 part by weight of alkyl aryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, the plants are inoculated with an aqueous spore suspension of the apple mildew pathogen Podosphaera leucotricha. The plants are then placed in a greenhouse at about 23°C and a relative atmospheric humidity of about 70%.

Evaluation is carried out 10 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

In this test, the compounds according to the invention of examples 1, 2, 3, 4, 5 and 6 showed, at an application rate of 100 g/ha, an efficacy of more than 90%.

Example B

Venturia - Test (Apple)/protective

Solvents: 24.5 parts by weight of acetone

24.5 parts by weight of dimethylacetamide

Emulsifier: 1 part by weight of alkyl aryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, the plants are inoculated with an aqueous conidia suspension of the apple scab pathogen Venturia inaequalis and then remain in an incubation cabin at about 20°C and 100% relative atmospheric humidity for 1 day.

The plants are then placed in a greenhouse at about 21°C and a relative atmospheric humidity of about 90%.

Evaluation is carried out 10 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

In this test, the compounds according to the invention of examples 1, 2, 3, 4, 5 and 6 showed, at in application rate of 100 g/ha, an efficacy of more than 90%.

Example C

Botrytis - Test (Bean)/protective

Solvents: 24.5 parts by weight of acetone

24.5 parts by weight of dimethylacetamide

Emulsifier: 1 part by weight of alkyl aryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, 2 small pieces of agar colonised by Botrytis cinerea are placed onto each leaf. The inoculated plants are placed in a dark chamber at about 20°C and 100% relative atmospheric humidity.

The size of the infected areas on the leaves is evaluated 2 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

In this test, the compounds according to the invention of examples 1, 2, 4 and 6 showed, at an application rate of 500 g/ha, an efficacy of more than 90%.

Example D

Sphaerotheca-Test (Cucumber)/protective

Solvent: 49 parts by weight of N, N-dimethylformamide

Emulsifier: 1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, then the concentrate is diluted with water to the desired concentration.

To test for protective activity, young cucumber plants are sprayed with the preparation of active compound at the stated application rate. 1 day after the treatment, the plants are inoculated with a spore suspension of Sphaerotheca fuliginea. The plants are then placed in a greenhouse at 70% relative atmospheric humidity and a temperature of 23°C.

Evaluation is carried out 7 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

In this test, the compounds according to the invention of examples 4, 15, 17 and 24 showed, at an application rate of 750 g/ha, an efficacy of more than 90%.